

CLAIMS

1. A cross-linked polyimide produced by polycondensing (a) tetramine(s), (a) tetracarboxylic dianhydride(s) and (an) aromatic diamine(s) in the presence of a catalyst, which cross-linked polyimide has a dielectric constant of not more than 2.7.
- 5 2. The polyimide according to claim 1, wherein said tetramine(s) is(are) (an) aromatic tetramine(s).
3. The polyimide according to claim 1, wherein said aromatic tetramine(s) is(are) at least one selected from the group consisting of bis(3,5-diaminobenzoyl)-1,4-piperazine, bis(3,5-diaminobenzoyl)-4,4'-diiminodiphenylether, bis-(3,5-diaminophenyl)-2,2'-dioxazol-4,4'-diphenylsulfone, bis(3,5-diaminophenyl)-2,2'-dioxazol-4,4'-biphenyl, 2,7-diamino-9,9'-(bis-4-aminophenyl)fluorene and bis(3,5-diaminobenzoyl)-1,4-diaminobenzene.
- 10 4. The polyimide according to any one of claims 1 to 3, which comprises a diaminosiloxane as a part of diamine component.
- 15 5. The polyimide according to any one of claims 1 to 4, which was produced by sequential reactions comprising polycondensing a tetramine, a tetracarboxylic dianhydride and an aromatic diamine in the presence of the catalyst to generate a polyimide oligomer, and then reacting the polyimide oligomer, a tetracarboxylic dianhydride and an aromatic diamine.
- 20 6. The polyimide according to claim 5, which was produced such that the difference between the number of moles of said tetracarboxylic dianhydride and the number of moles of said aromatic diamine, which are reacted with said tetramine is 2 moles per 1 mole of said tetramine.
- 25 7. The polyimide according to claim 6, which was produced by a process comprising polycondensing said tetramine, 4 moles of said tetracarboxylic dianhydride and 4 moles of said aromatic diamine per 1 mole of said tetramine to generate said polyimide oligomer, and then reacting the polyimide oligomer, 4 moles

of the tetracarboxylic dianhydride and 2 moles of the aromatic diamine.

8. The polyimide according to claim 6, which was produced by a process comprising polycondensing said tetramine, 8 moles of said tetracarboxylic dianhydride and 4 moles of said aromatic diamine per 1 mole of said tetramine to generate said polyimide oligomer, and then reacting the polyimide oligomer, 2 moles of tetracarboxylic dianhydride and 4 moles of aromatic diamine.

5 9. The polyimide according to any one of claims 1 to 8, which has a weight average molecular weight based on polystyrene of 15,000 to 300,000.

10 10. The polyimide according to any one of claims 1 to 9, which has a dielectric constant of 1.9 to 2.2.

11. A process for producing a composition containing a cross-linked polyimide, comprising polycondensing (a) tetramine(s), (a) tetracarboxylic dianhydride(s) and (an) aromatic diamine(s) in a polar solvent containing toluene or xylene in the presence of a catalyst under heat.

15 12. The process according to claim 11, wherein said tetramine(s) is(are) (an) aromatic tetramine(s).

13. The process according to claim 12, wherein said aromatic tetramine(s) is(are) at least one selected from the group consisting of bis(3,5-diaminobenzoyl)-1,4-piperazine, bis(3,5-diaminobenzoyl)-4,4'-diiminodiphenylether, bis-(3,5-diaminophenyl)-2,2'-dioxazol-4,4'-diphenylsulfone, bis(3,5-diaminophenyl)-2,2'-dioxazol-4,4'-biphenyl, 2,7-diamino-9,9'-(bis-4-aminophenyl)fluorene and bis(3,5-diaminobenzoyl)-1,4-diaminobenzene.

20 14. The process according to any one of claims 11 to 13, wherein a diaminosiloxane is contained as a part of diamine component.

25 15. The process according to any one of claims 11 to 14, wherein said catalyst is a binary catalyst comprising (an) acid(s) selected from the group consisting of oxalic acid, malonic acid, formic acid and pyruvic acid, and a base, or a binary catalyst

comprising a lactone and a base.

16. The process according to claim 15, wherein said catalyst is a binary catalyst comprising oxalic acid and a base, or a binary catalyst comprising a lactone and a base.

5 17. The process according to claim 16, wherein the reactants are directly imidized in the presence of said binary catalyst at 160°C to 200°C.

18. The process according to any one of claims 11 to 17, by sequential reactions, comprising polycondensing a tetramine, a tetracarboxylic dianhydride and an aromatic diamine in the presence of the catalyst to generate a polyimide oligomer,
10 and then reacting the polyimide oligomer, a tetracarboxylic dianhydride and an aromatic diamine.

15 19. The process according to claim 17, wherein the difference between the number of moles of said tetracarboxylic dianhydride and the number of moles of said aromatic diamine, which are reacted with said tetramine is 2 moles per 1 mole of said tetramine.

20. The process according to claim 19, wherein said tetramine, 4 moles of said tetracarboxylic dianhydride and 4 moles of said aromatic diamine are reacted per 1 mole of said tetramine to generate said polyimide oligomer, and then reacting the polyimide oligomer, 4 moles of the tetracarboxylic dianhydride and 2 moles of the aromatic diamine.

25 21. The process according to claim 19, wherein said tetramine, 8 moles of said tetracarboxylic dianhydride and 4 moles of said aromatic diamine are reacted per 1 mole of said tetramine to generate said polyimide oligomer, and then reacting the polyimide oligomer, 2 moles of the tetracarboxylic dianhydride and 4 moles of the aromatic diamine.

22. A process for producing a cross-linked polyimide composition, comprising adding (a) tetracarboxylic dianhydride(s) and (an) aromatic diamine(s) to the

polyimide composition produced by the process according to any one of claims 11 to 21, mixing the mixture and polycondensing them.

23. A process for producing a cross-linked polyimide composition, comprising carrying out said process according to any one of claims 11 to 21, in a linear polyimide composition produced by the same process as the process according to claim 11 except that said tetramine(s) is(are) not used.

5 24. A cross-linked polyimide composition produced by the process according to any one of claims 11 to 22.

10 25. The polyimide composition according to claim 24, wherein the cross-linked polyimide in said polyimide composition has a weight average molecular weight based on polystyrene of 15,000 to 300,000.

26. The cross-linked polyimide composition according to claim 24 or 25, further comprising a linear polyimide produced by the same process as the process according to claim 11 except that said tetramine is not used, and which composition is in the form of liquid at room temperature.

15 27. A photosensitive cross-linked polyimide composition, further comprising a photoacid generator in said composition according to any one of claims 24 to 26.

28. A process for producing a patterned polyimide film, comprising casting a solution of said photosensitive cross-linked polyimide composition according to claim 27 on a substrate, heating the cast composition at 60°C to 90°C to obtain a film, irradiating the film through a mask, and etching the resultant with an alkaline solution to form a positive image.

20 29. The patterned polyimide film produced by the process according to claim 28.

30. An electrical or electronic equipment or a part thereof, which comprises an insulation material, insulating substrate or protection material, that contains said 25 cross-linked polyimide according to any one of claims 1 to 10.

31. The electrical or electronic equipment or a part thereof according to claim 30,

wherein said cross-linked polyimide is used as (1) an interlayer insulation film between semiconductor elements, (2) a laminate sheet, multilayer circuit substrate or a substrate of a flexible copper-clad plate, or (3) a semiconductor chip-coating film.

32. The electrical or electronic equipment or a part thereof according to claim 31,
5 wherein said semiconductor chip-coating film is a passivation film, α -ray-shielding film or buffer coat film.

33. The electrical or electronic equipment or a part thereof according to any one of claim 30 to 32, wherein said cross-linked polyimide is a positive-type photosensitive polyimide containing a photoacid generator, and wherein said
10 insulation material or protection material is formed by photolithography.

34. The electrical or electronic equipment or a part thereof according to any one of claim 30 to 32, wherein said insulation material or protection material is formed by screen printing.

35. The electrical or electronic equipment or a part thereof according to any one of claim 30 to 32, wherein said cross-linked polyimide comprises anionic group-containing units, and wherein said insulation material or protection material is formed by electrodeposition.
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36. The electrical or electronic equipment or a part thereof according to claim 35, wherein group which becomes an anion in aqueous solution is carboxylic group or a salt thereof.
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